LCA Case Studies

Selection of a Remediation Scenario for a Diesel-Contaminated Site Using LCA*

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Abstract

Goal and Scope. A comparison of in situ and ex situ treatment scenarios for a diesel-contaminated site was performed using an evolutive LCA. Treatment time along with primary (residual contamination left in soil or groundwater after treatment) and secondary (impacts due to remediation) environmental impacts were considered. The site under study had a Light Non Aqueous Phase Liquid (LNAPL) thickness of up to 1 m, a diesel soil concentration of 10,500 mg/kg and a residual contamination in groundwater.

Methods. Four treatment scenarios to remove LNAPL and to treat soil and groundwater were compared: 1) pump and treat 2) bioslurping, bioventing and biosparging 3) bioslurping, bioventing and chemical oxidation and 4) ex situ treatment using biopiles. The technologies' design was performed using simulation tools and analytical equations. The LCA was evaluated for each year of treatment. Environmental impacts were assessed using the U.S. EPA Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI) method.

Results and Discussion. The biological in situ scenario (2) showed the least primary and secondary impacts but its treatment time was more than 4 times longer than that obtained for the ex situ scenario (4). The ex situ scenario showed the best treatment time but its secondary impacts were significantly higher than those found for the biological in situ scenario due to the pavement of the treatment area. The combined biological and chemical in situ scenario (3) was the worst in terms of secondary impacts while the pump and treat scenario (1) was the worst in terms of primary impacts. Two scenarios were selected: one based upon low environmental impacts and the other on the fastest treatment time.

Conclusions. Even without excavation, an in situ treatment can generate more secondary impacts than an ex situ treatment. Low environmental impact scenarios require time while rapid treatment scenarios generate high environmental impacts. The selection of the best remediation scenario will depend on the site owner's priority.

Recommendations. Better characterization factors for aggregated substances are required.

Keywords: Contaminated soil; contaminated groundwater; diesel; life cycle assessment; primary impact; secondary impact; TRACI method; treatment time; ex situ remediation; in situ remediation

Introduction

Life cycle assessment (LCA) has been described as a good decision-making tool for managers concerned with choosing the best product or service from an environmental point of view. In the site remediation field, LCA can help in choosing the best technology to reduce the environmental burden. While several studies have been conducted on the subject, none have managed to provide the best treatment scenario for a site where both soil and groundwater are contaminated. Moreover, most of these studies covered ex situ technologies at the expense of in situ ones. Past studies mostly analyzed ex situ soil treatment technologies. Page et al. [1] conducted an LCA for site remediation on a lead-contaminated site using excavation and disposal. Diamond et al. [2] developed an LCA framework: a qualitative method to investigate remediation activities. Their article cited examples qualitatively comparing the following technologies: bioattenuation (no action), encapsulation, excavation and disposal, vapor extraction, in situ bioremediation and soil washing. Some authors have compared soil washing, biopiles, thermal treatment and capping for PAH contamination [3], while others have looked at disposal on secure landfills, on-site containment, bio-leaching and liming stabilization for sulfur contamination [4]. These case studies were conducted to show that the LCA method is effective for specific sites for which different choices are available financially, legally and technically and where the final decision can consider the environment a priority [3,4]. A detailed analysis of the impacts of a diesel-contaminated soil treated in biopile has also been performed [5]. One of the first LCA studies undertaken on site remediation pertained to the development of a tool to calculate and discuss environmental impacts of soil and groundwater treatment. However, it only compared hydrocarbon-contaminated groundwater pump and treat technologies [6]. While one detailed impact comparison assessment (LCIA) of in situ and ex situ technologies has been introduced (natural attenuation, off-site confinement and thermal treatment of spent pot lining contaminated soil) [7], none has been conducted on a site where both soil and groundwater have been contaminated. Moreover, most of these studies have only compared ex situ technologies where the major impact was soil transport [1,4,5,7,8]. A literature survey reveals no in depth evaluation has been detailed for cases where both in situ and ex situ technologies have been applied to site remediation.

*** This paper is openly accessible!**

The remediation of a contaminated site is characterized by its primary and secondary impacts but most authors who have performed LCAs have only evaluated the secondary impacts (related to remediation treatment) [1–4,6]. Others have tried to evaluate the primary impacts (related to residual contamination) by attributing a characterization factor (CF) to the contaminant [5,7,8]. It has been demonstrated that primary impacts should not be neglected because they can represent up to 92% of global impacts [5]. These impacts can clearly be used to increase the LCA significance for site remediation [7] further studies are, however, needed to confirm this.

Aside from this, very few studies have detailed the time factor in LCAs: some have conducted a 25-year study [2] while others have used a 50-year period [6]. The time factor is crucial in site remediation, because depending on the contaminant and its concentration, some technologies will require hours and others, years to reach a treatment target. None of the surveyed studies comparing different technologies took the treatment time of each technology or even their efficiency into account.

The goal of the present study is to compare the environmental performance of four treatment scenarios considering both in situ and ex situ technologies. This comparison is based on the treatment time necessary to reach Quebec's B criterion in soil (700 $mgkg^{-1}$) and clean groundwater (0.1 mgL^{-1}) and on the magnitude of primary and secondary impacts in order to select the best technologies. A diesel-contaminated site where both soil and groundwater were impacted was modeled to obtain accurate data on the technologies used.

1 Methodology

1.1 Case study definition

This study was performed on a site where a 375 m^3 diesel tank spill occurred. The contaminated area was located 600 m from a river shore in the northern part of the province of Quebec, Canada. The spill covered an area of 480 m^2 in the vadose zone and reached the groundwater located 6 m below the surface. The diesel spanned over an area of 2700 m^2 on top of the groundwater and its thickness varied from 0.1 m to 1 m.

The soil type was mainly medium sand and the diesel concentration in the vadose zone (soil above the groundwater level) was approximated to be 10,500 $mgkg^{-1}$. The decontamination was necessary in this case in order to stop or slow down the diesel migration towards the river. Due to the presence of light non aqueous phase liquid (LNAPL), it was necessary to remove it prior to using any other technology to clean the soil or groundwater. Four scenarios were developed in order to compare in situ and ex situ technologies, with different efficiencies to remediate this site. **Table 1** shows the different scenarios and chosen technologies.

The first scenario included a traditional in situ treatment with ex situ groundwater treatment, the second consisted of an in situ biological treatment, the third included a combined in situ biological and chemical treatment and the last, an ex situ soil treatment with groundwater monitoring. The latter scenario was conducted on-site in order to avoid impacts related to contaminated and clean soil transport. Indeed, these impacts were shown to be most significant in other studies [1,4,5,7,8].

1.2 LCA goal and scope

In the present study, the function was to restore a contaminated site and the functional unit was defined as the remediation of a 375 m^3 diesel-contaminated site to the Quebec B criterion in soil (700 $mgkg^{-1}$) and to the detectable limit of C_{10}–C_{50} for potable, groundwater and surface water (0.1 mgL^{-1}) [9]. The reference flow was established as a site. The system boundary did not include site characterization. A schematic view of the system boundaries is presented in **Fig. 1**. In site remediation, the LNAPL phase must first be removed. The vadose and saturated zone treatments follow simultaneously. The site preparation must be done at the beginning of each treatment and all the equipment must be removed at the end. Since the study focuses on the treatments' impacts throughout the years, which it is why an evolutive LCA is used, each studied year included the previous year's impacts. The site preparation included the production and transport of all needed equipment and materials for each technology. The production of non road equipment (e.g. bulldozer) was not included in the system since it can be reused elsewhere. Furthermore, the dismantling phase included the machinery return transport and clean soil transport when needed. Clean soils were also needed to fill up the wells.

Since the LCA performed does not compare specific products but management scenarios for site remediation treatment, the ISO standards recommendations for products comparison were not entirely followed. For instance, normalized data were used to compare the scenarios.

1.3 Technology design

All technologies were simulated either theoretically or numerically with commercial software. These simulations helped determining the treatment time and the necessary equipment to be used in the life cycle inventory (LCI) for each technology. Moreover, the simulations helped to quantify in time the groundwater contaminants' migration and concentration. Since modeling contaminant distribution in groundwater requires good quality and site-specific data on a wide range of hydrogeological properties, a conceptual model was developed as the basis for numerical modeling of groundwater

Table 1: Technologies included in each restoration scenario

	Scenario 1	Scenario 2	Scenario 3	Scenario 4
LNAPL removal	Oil removal	Bioslurping	Bioslurping	Bioslurping
Soil treatment	Natural attenuation	Bioventing	Bioventing	Excavation with on site biopiles
Groundwater treatment	Pump and treat	Biosparging	Chemical Oxidation	Natural attenuation

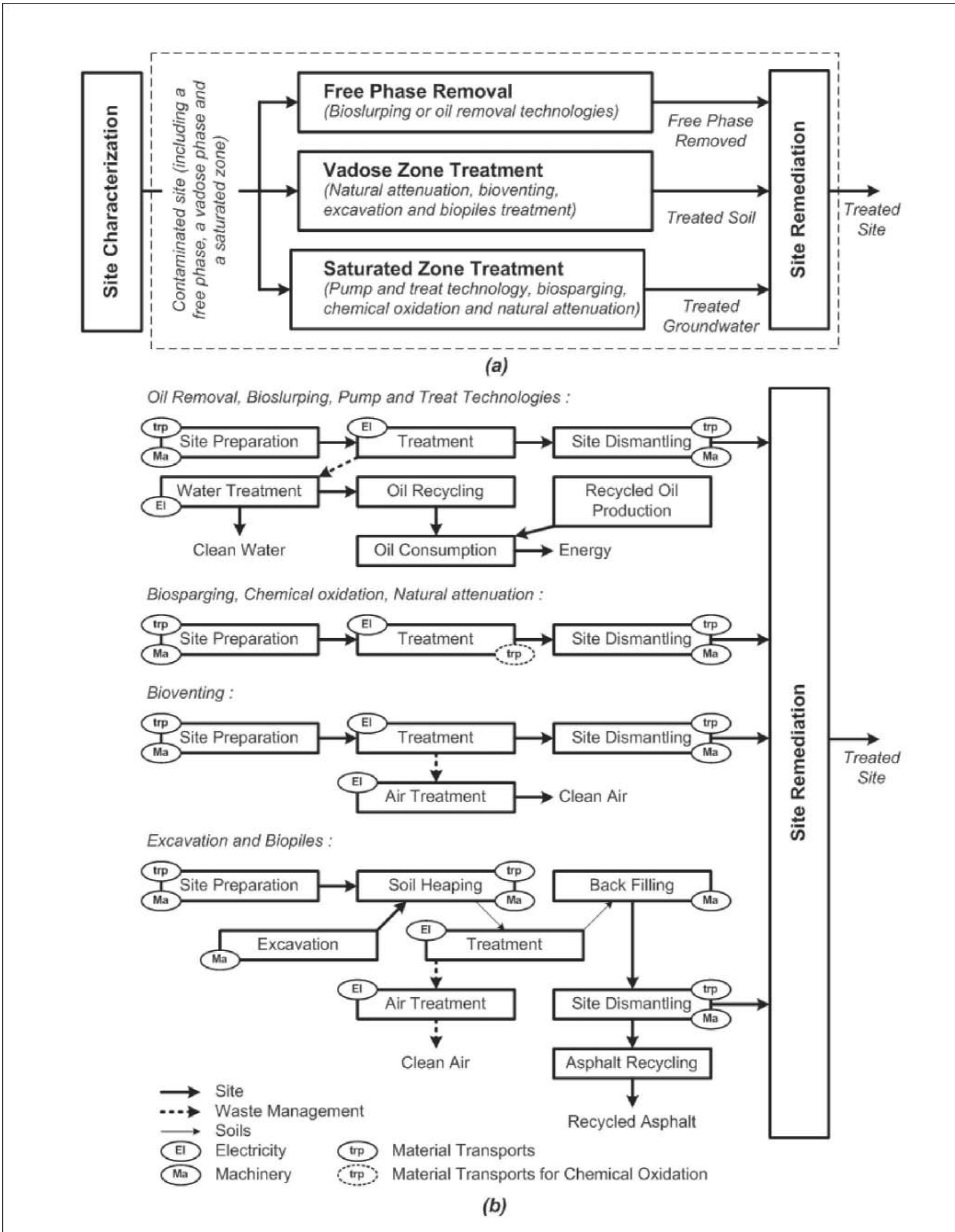

Fig. 1: a) System boundary b) Flow diagram for each technology

Table 2: Material requirements and treatment time for each technology

Technologies	Description
LNAPL Phase Removal	
Oil Removal	• 6-wells 15.24 cm diameter, 9 m deep and spaced by 10 m • flow rate: 10 $m^3d^{-1}well^{-1}$ • 4 monitoring wells 5.1 cm diameter, 9 m deep • 6 pumps, 3 compressors 2.2 kW • 6.1 m site trailer • oil/water separator 0.4 kW • 13 189 L drums of activated carbon • 9.1 m^3 oil reservoir • Treatment time: 2 years with 6 months off
Bioslurping	• 54 wells 5.1 cm diameter 9 m deep on 6 lines, 3 lines spaced by 5 m and 3 others by 10 m • liquid flow rate: 13.5 $m^3d^{-1}well^{-1}$ • 4 monitoring wells 20.3 cm diameter, 5.4 m deep with O_2 and CO_2 captors • 1 pump 22.4 kW • 6.1 m site trailer • 13 70 L drums of activated carbon • air/liquid separator 0.4 kW • oil/water separator 0.4 kW • 3 9.1 m^3 oil reservoirs • 3.2 m^3 biofilter • 55 L moisture tower • Treatment time: 2 years with 6 months off
Vadose Zone: Soil Treatment	
Natural Attenuation	• 2 wells, 20.3 cm diameter, 5.5 m deep with O_2 and CO_2 captors, tested 3 times a year • Soil sampling every 2 years • diesel water solubility: 5 mgL^{-1} [31] • Monitoring time: 300 years
Bioventing	• 2 air extraction wells, 5.1 cm diameter, 5.5 m deep, spaced by 17.5 m • biological influence radius: 12.3 m • extraction suction: 7 cfm/wells • 4 monitoring wells, 20.3 cm dia. 5.5 m deep • 1 blower 300 W, 1 pump 375 W • 1.4 m^3 biofilter (4 media changes) • 45 L moisture tower • Biodegradation rate (k_B): 3.29 $mgkg^{-1}$ [11] • Treatment time: 8.2 years continually • Restoration up to QC B criterion 700 $mgkg^{-1}$
Excavation & Biopile treatment	• 19 biopiles of 531 m^3 of soil (5 biopiles per year) • blower 2.6 kW and pump 0.4 kW • total extraction air flow rate for 5 biopiles: 374 m^3h^{-1} • paved area: 3650 m^2 • 220 L moisture tower • 21 m^3 biofilter • steel fence (perimeter 218 m) • 5 LDPE semi–permeable geotextiles of 800 m^2 • 6.1 m site trailer • Oxygen utilization rate: 88% d^{-1} [33] • Biodegradation rate: 48.33 $mgkg^{-1}$ • Diesel concentration in soil: 13 225 $mgkg^{-1}$ (LNAPL withdrawn with excavated soil) • Restoration up to QC B criterion 700 $mgkg^{-1}$ • Treatment time: 3.75 years (3 months off between treatments)
Saturated Zone: Groundwater Treatment	
Pump & Treat	• 3 wells, 15.2 cm diameter, spaced by 30 m, 14 m deep • 4 monitoring wells 5.1 cm dia. , 9 m deep • pumping rate: 130 $m^3d^{-1}well^{-1}$ • 26 drums of 189 L activated carbon per year • 6.1 m site trailer • oil/water separator 0.4 kW • 9.1 m^3 oil reservoir • 3 pumps of 130 $m^3d^{-1}well^{-1}$, 3 compressors of 2.2 kW (lifetime 15 years) • Treatment time: 300 years continually • Final concentration in groundwater: 0.1 mgL^{-1}
Biosparging	• 84 injection wells, 5.1 cm dia. 9 m deep: 81 in the source in 9 lines spaced by 8 m and 3 in a line at 80 m downstream of the source spaced by 16 m and shut down after 5 years. • 8 monitoring wells, 5.1 cm dia. 9 m deep • Influence radius of injection wells: 6 m • Oxygen rate transfer between air and water: 2% • Rate of O_2 injected transferred to groundwater: 0.035 $kgd^{-1}well^{-1}$ • First order biodegradation constant: 0.05 d^{-1} [32] • Injected air flow rate: 6 $m^3d^{-1}well^{-1}$ • Operation: 180 days per year • 4 compressors 1.5 kW • 6.1 m site trailer • Treatment time: 36 years • Final concentration in groundwater: 0.1 mgL^{-1}
Chemical Oxidation	• 125 steel wells 5.1 cm dia. 9 m deep, in 10 lines spaced by 5 m • 8 monitoring wells, 5.1 cm dia. 9 m deep • Influence radius of injection wells: 3 m • 3 Pumps: 1.5 kW, 0.2 kW, 0.1 kW • Air compressor 1.5 kW • CH_3COOH 50 % wt. (lower pH to 4) • Oxidant: Fenton reagent (H_2O_2 50 % wt. and Fe_2SO_4 dry) • 2 2 m^3 mixing reservoirs 0.4 kW • Treatment time: 3.7 years • Final concentration in groundwater: 0.1 mgL^{-1}
Natural Attenuation	• 6 monitoring wells, 15.2 m dia. 9 m deep tested 4 times a year • Monitoring time: 6 years • Naphthalene properties used for the simulation • (most soluble substance in diesel)

flow and contaminant transport by performing a synthesis of existing data through available reports. The decrease in contaminant due to treatment was mostly modelized as linear in time. **Table 2** presents all the design data including the peripheral equipment of each technology used for the LCI.

Two technologies were designed to remove the LNAPL phase: oil removal and bioslurping. Treatment was ended when all residual LNAPL was considered to be absorbed. This period was estimated at 2 years. Oil removal consists of creating a drawdown of the groundwater towards the pumping well at the interface of the vadose and saturated zones; a pump located into the well is then able to recuperate the LNAPL. The bioslurping technology consists of sucking up the LNAPL at the vadose and saturated zones' interface without using a drawdown of the groundwater. The air suction creates an oxygen inflow in the soil stimulating contaminant biodegradation. The LNAPL migration and recuperation for both technologies were simulated with the

Compflow software [10]. The model range covered a 60 m zone in radius around the spill zone .

Three technologies were chosen to remediate the vadose zone: natural attenuation, bioventing and biopiles. Natural attenuation can be achieved with natural phenomenon like dilution, dispersion, sorption, volatilization and biodegradation. This process requires, however, long-term monitoring of the contaminant plume. The contaminant dilution by rainwater was solely used to estimate the natural attenuation of diesel in the vadose zone. The hypothesis was that all the recharging water (400 $mmyear^{-1}$) from the precipitation would dissolve the diesel. The monitoring time was the same as the treatment time for the pump and treat technology. Bioventing consists of extracting air from the soil in order to create an oxygen inflow to increase contaminant biodegradation. The extracted air is then treated with a biofilter. The bioventing design was based on Leeson and Hinchee's model [11]. Because no hydrocarbon biodegradation rate (k_B) was available from the site, an average value from 145 sites was used (3.29 $mgkg^{-1}$) [12]. Soil treatment in biopiles consists of increasing the contaminant biodegradation conditions by extracting air and adding nutriments. The extracted air is also treated with a biofilter. The ex situ biopile treatment technology was designed to treat the entire contamination source. All contaminated soil in the vadose and saturated zone was excavated (7,920 m^3) and treated.

Four technologies were selected to treat groundwater: pump and treat, biosparging, chemical oxidation and natural attenuation. The pump and treat technology consists of pumping the groundwater and treating it with an activated carbon filter. The amount of water needed to dissolve the residual contamination was estimated (4.2e7 m^3). By setting the pumping rate (130 $m^3d^{-1}well^{-1}$), the calculated influence radius (15 m) determined the number of required wells (3) and treatment time (300 years). Biosparging consists of injecting air into the groundwater in order to increase contaminant biodegradation. The immiscible hydrocarbon phase biodegradation was simulated with the Bionapl model [13]. Setting the influence radius of biosparging (6 m) allowed for calculation of the required injection wells (84) and treatment time (36 years). The in situ chemical oxidation technology consists of injecting an oxidant into the groundwater in order to oxidize the contaminant. Fenton reagent (H_2O_2 and $FeSO_4$) was chosen for the chemical oxidation design because of its known efficiency with hydrocarbon products [14,15]. The hypothesis made to estimate the required quantity of hydrogen peroxide and iron sulfate was that the oxidized substance was $C_{14}H_{30}$ because of its high concentration in diesel [16]. **Reactions (1)** and **(2)** were used to evaluate the quantity of needed H_2O_2 and $FeSO_4$.

$$C_{14}H_{30} + 86\ OH\cdot \rightarrow 14\ CO_2 + 58\ H_2O \qquad (1)$$

$$86\ Fe^{2+} + 86\ H_2O_2 \rightarrow 86\ OH\cdot + 86\ Fe^{3+} + 86\ OH^- \qquad (2)$$

Natural attenuation in the saturated zone is also a long-term monitoring process. The aqueous phase contamination migration, for the natural attenuation in the saturated zone, was simulated by coupling the Modflow and RT3D models [17,18]. Modflow simulated groundwater runoff while RT3D simulated the aqueous phase contaminant transport. Only the dilution and dispersion of contaminant where used by the models. The monitoring time (6 years) corresponds to the plume's time to reach the surface water. The diesel concentration in groundwater reaching the river was estimated at 1 mgL^{-1}.

1.4 Life cycle inventory (LCI)

For each of the four scenarios, the environmental load was calculated in relation to the functional unit. The needed equipment, diesel and electricity quantities were also calculated in relation to the treatment time for each technology. Only secondary data from the Franklin and Ecoinvent databases were used [19,20] to complete the LCI. The LCI modeling was performed with Microsoft ® Excel [21] and Matlab ® [22]. No LCA commercial software package was used in this study in order to understand every step of the analysis. The basic calculations followed a set of three equations: $As = f$; $g = Bs$ and $s = A^{-1}f$, where A is the technology matrix, A^{-1} its inverse, s a scaling vector, f the final demand vector, g the inventory vector and B the intervention matrix [23]. To limit the volume of treated data, every LCA was based on a set of determined hypotheses. The main assumptions used in this LCI elaboration were the following:

- The duration of LNAPL removal from the period covering site preparation to site dismantling was two years (including a 195-day shutdown period during winter).
- The electricity data was obtained from the Ecoinvent database using the Quebec Power Grid Mix [24] (95.05% hydropower; 0.55% hard coal; 0.11% oil; 0.35% natural gas; 3.09% nuclear; 0.76% peat and 0.09% wind power).
- All transportations to and from the site were included (equipment, samples for analysis and clean soil). Distances varied depending on the best supplier for a technology. Truck emissions were calculated using the appropriate factors for the type of vehicle, fuel type and load. Machinery emissions were calculated using the NON ROAD model [25,26].
- All recovered diesel was recycled but its transport to a recycling unit was not included.
- All treated water recuperated from bioslurping, oil removal or pump and treat was sent to the nearest storm sewer.
- No site dismantling is required for natural attenuation in the vadose zone, the monitoring wells are left in place because they are sealed.
- Due to the high environmental impact of asphalt production, the asphalt paving was deliberately restricted to the contaminant-sensitive parts of the treatment area: the foundation of the biopiles and the storage area (3,650 m^2) [5].
- Virgin asphalt was used for the pavement of the biopile treatment area but all the removed asphalt was recycled. The asphalt recycling process was included in the system but the transport of the recovered asphalt was not.
- Ex situ monitoring activities like laboratory analyses were not taken into account due to their minor role [2], however, as indicated before, sample transport to the laboratory was included.
- All staff activities were excluded from this study.

1.5 Life cycle impact assessment (LCIA)

The classification and characterization of impacts were performed using the US EPA TRACI Method [27]. Since the TRACI method offers no normalization factors (NFs), Canadian NFs had to be calculated. NFs were needed to sum all impacts from each category in order to compare primary and secondary impacts. NFs were calculated using the Impact 2002+ method [28]. Nine relevant environmental impact categories were considered: global warming, ozone depletion, acidification, eutrophication, photochemical smog, ecotoxicology, human health cancer effects (HHC), human health non cancer effects (HHNC) and human health criteria. When no characterization factor (CF) existed for an aggregated substance (a substance that is a mix of several pure substances), the minimum and maximum factors of the pure substances included in the aggregated substance were used. This method was chosen to cover both optimistic and worst case scenarios. The minimum and maximum impact values are presented with bars on the graphs. Unfortunately, the TRACI method does not include soil CFs. The reason is that all the substances in soil are assumed to eventually end up in water [29]. Therefore, the TRACI aquatic CFs were used to characterize soil emissions (data from the Ecoinvent database). The same method was applied to determine the primary impacts of the diesel (an aggregated substance) contamination in soil and water before and after treatment. The primary impacts were calculated by multiplying the residual quantity of diesel in soil and water by its aquatic min. and max. CFs. It is important to note that the variability in diesel CFs can yield a wide range in the final LCA quantitative results.

2 Results and Discussion

To get a better view of each technology, the comparison was conducted between technologies used in the same zone instead of comparing the four scenarios. This way, the preferable technology for each zone can be determined to remediate a contaminated site and to finally create the best scenario.

2.1 Comparison of LNAPL removal technologies

Table 3 presents the input materials and output emissions for both technologies. In order to lighten the table, note that the output emissions presented here correspond to 5% or more of all emissions.

Bioslurping always has the most input materials and output emissions except for activated carbon, clean soils and solid wastes. The oil removal technology pumps more water while removing LNAPL so it needs more activated carbon to treat a larger flow rate which in turn creates more solid wastes. It also needs more clean soil to fill up its wells even if there are fewer of them (6) than there are for bioslurping (54) because they are three times larger. The analysis of these technologies also shows that recycling diesel can restrain the dissolved solids leading them to end up in water emissions while fossil CO_2 end up in air emissions (negative output emissions). Also, the important substances found in water and soil emissions were mostly non toxic (only unspecified oils were identified as toxic).

Table 3: Mass input materials and mass output emissions for the LNAPL removal phase

Materials (inputs)		Oil removal	Bioslurping
Diesel $_{machinery}$ (m^3)		3.05	5.4
Diesel $_{transport}$ (m^3)		1.8	1.62
Cement (kg)		378.3	1,059
Bentonite (kg)		742.7	1,011
Sand (kg)		1,583	2,514.5
Gravel (kg)		0	62.5
PVC (kg)		545.4	967
HDPE (kg)		289	1,044
Activated carbon (kg)		884.5	221
Biofilter (kg)		0	617
Electricity (GJ)		79.8	695.6
Clean soil (kg)		2,846	1,951
Emissions (outputs)			
Biogenic CO_2 (kg)	Air	596.6	1,495
Fossil CO_2 (kg)		–3,984	–14,926
Calcium (kg)	Water	4.1	21.9
Chloride (kg)		23.1	48.1
Dissolved solids (kg)		–257	–566.4
Unspecified oils (kg)		6.36	34
Silicon (kg)		6.7	44.4
Sodium (kg)		19.8	38
Sulfate (kg)		–2.57	13.1
Calcium	Soil	0.07	0.06
Chloride (kg)		0.08	0.24
Unspecified oils (kg)		0.58	1.53
Solid waste (kg)		730.6	670
Removed diesel (m^3)		67	144
Water (m^3)		19,800	10,110

Comparing the secondary impacts for each impact category, **Table 4** shows that oil removal has less impact than bioslurping for each category. The difference is mostly due to well installation. Note that the most significant categories for both technologies are water and soil ecotoxicity. The

Table 4: Total average normalized impacts by impact categories for the oil removal stage

Categories (person*year)	Media	Oil removal	Bioslurping
Global warming	Air	–0.002	–0.008
Ozone depletion	Air	0.003	0.013
Acidification	Air	0.78	1.13
Eutrophication	Air	1.26	1.48
	Water	0.016	0.11
	Soil	0.005	0.003
Photochemical smog	Air	1.38	1.64
Ecotoxicity	Air	0.057	0.53
	Water	**1.3 E 3**	**6.8 E 3**
	Soil	**1.2 E 2**	**3.1 E 2**
Human health cancer	Air	1.2 E-8	7.4 E-8
	Water	4.2 E-6	2.3 E-5
	Soil	3.9 E-7	1.0 E-6
Human health criteria	Air	1.1	3.27
Human health non cancer	Air	5.0 E-4	0.26
	Water	–1.2 E-5	3.4 E-3
	Soil	5.2 E-5	1.4 E-4

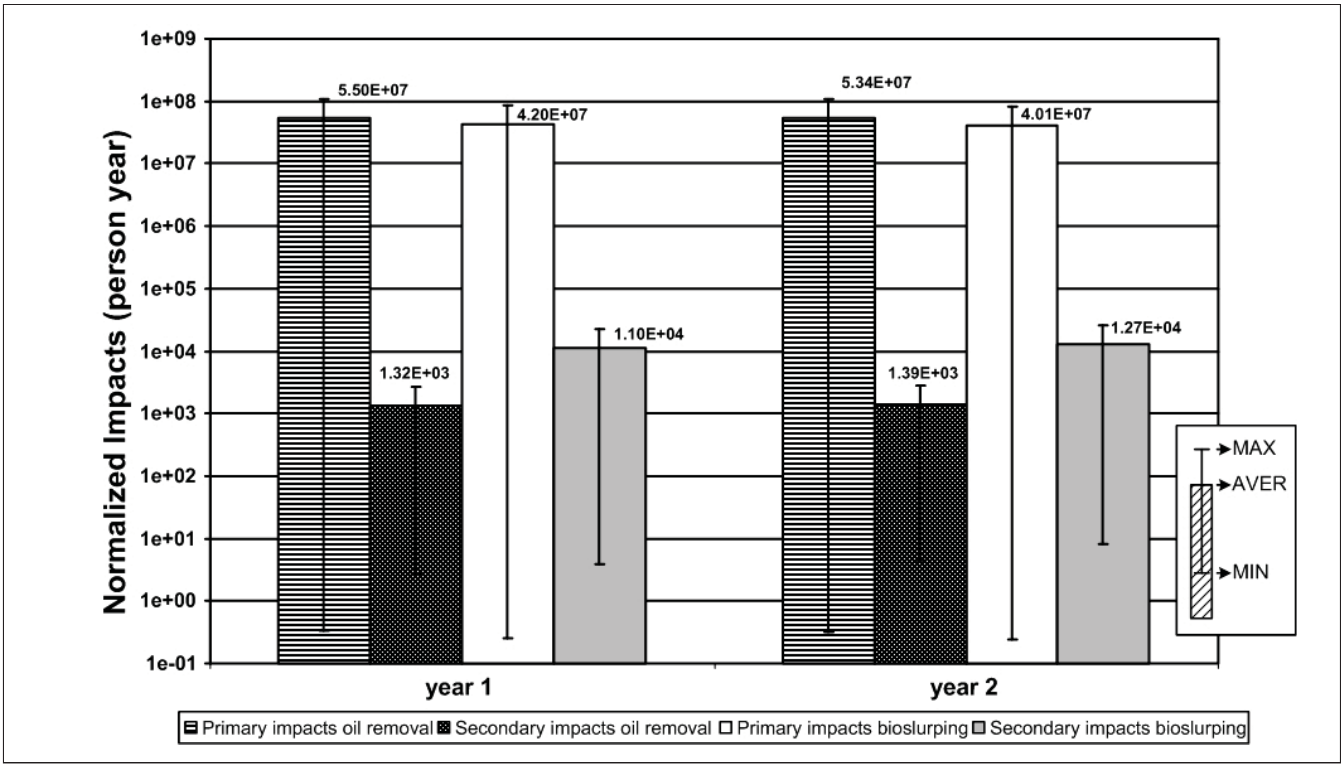

Fig. 2: Comparison of the normalized primary and secondary impacts for the oil removal technologies over the years; the values indicate the average normalized impacts

substance contributing the most to these categories is unspecified oils from the diesel and electricity production. Note also that since unspecified oils are an aggregated substance, their characterization factor in the analysis had the same minimum and maximum factors as diesel.

From a primary impacts standpoint, **Fig. 2** indicates that bioslurping (4.01e7 pers.year) appears better than oil removal (5.34e7 pers.year) because it actually removes more diesel (145m^3 vs 67m^3). For both technologies, most of the secondary impacts occurred the first year due to material production and installation. Moreover, most of the LNAPL was also recovered during the first year of treatment. This conclusion is obtained when comparing the primary impacts of the first and second year of treatment.

2.2 Comparison of soil treatment technologies

The comparison of soil treatment technologies (natural attenuation (NA), bioventing and biopile) can first be conducted based on their treatment time. The monitoring of NA was estimated at 300 years, the biopile required 4 years of treatment while soil bioventing required 8. The two active technologies treated the soils down to 700 mgkg^{-1}.

According to the Quebec Regulation on Contaminated Soils (QRCS), this concentration is acceptable for a residential and commercial zone (B criterion). NA, however, left 61.1 m^3 of diesel in the soils even after three centuries. Based on the contaminated soil volume (2,880 m^3), this represents an estimated diesel concentration of 9,878 mgkg^{-1} which is almost 3 times the QRCS regulated value for an industrial site (3,500 mgkg^{-1} hydrocarbon content).

Comparison of material inputs (**Table 5**) shows that NA used the least materials while biopiles used the most. The principal mass inputs for biopiles were asphalt (501 ton) and gravel (1,788 ton) needed to pave the biopile treatment area. This result is comparable with those of Toffoletto et al. [5]. When it comes to output emissions, NA produced more fossil CO_2 emissions than bioventing and it produced more biogenic CO_2 emissions than NA. The major production of fossil CO_2 emissions is from transports which are more numerous for NA (annual samplings). The major production of biogenic CO_2 emissions is from electricity production (8 years of continuous use to power a blower and a pump). The biopile emissions are significantly higher than those produced by NA and bioventing put together. It is important to note that the biopile site preparation stage was the most significant in terms of emissions. Paving was the most influential activity for that stage, once again due to the production and transport of materials. This result is also comparable to those of Toffoletto et al [5].

When the average normalized secondary impacts (ANSIs) are compared by categories (**Table 6**), the biopile treatment generates more impacts than any other treatment. NA and bioventing show, for their part, similar ANSIs. Bioventing has more ANSIs only for water ecotoxicity (1.3e3 pers.year) and water human health cancer effects (4.4e-6 pers.year). The most significant impact categories are water and soil ecotoxicity for the three technologies. The most influential

Table 5: Mass input materials and mass output emissions for the soil treatment phase

Materials (inputs)		Natural attenuation	Bioventing	Excavation & Biopile
Diesel $_{machinery}$ (m^3)		0.14	0.38	46
Diesel $_{transport}$ (m^3)		0.85	0.5	1.7
Cement (kg)		0	12.6	–
Bentonite (kg)		301.6	612	–
Sand (kg)		70.1	218	–
Gravel (kg)		31.3	62.5	1,788 t
Asphalt (t)		–	–	501
PVC (kg)		–	199	4,368
HDPE (kg)		–	79	783
LDPE (kg)		–	–	2,944
Steel (kg)		–	–	2,802
Biofilter (kg)		–	1,015	3,213
Electricity (GJ)		–	174	268
Clean soil (kg)		–	93	–
Latex (kg)		46.8	1.4	5
Glass (kg)		3,600	108	32.5
Wood chips (t)		–	–	317
Urea (kg)		–	–	16,118
Diammonium phosphate (kg)		–	–	3,521
Emissions (outputs)				
Biogenic CO_2 (kg)	Air	173	817.3	2971
Fossil CO_2 (kg)		6,143	4,543	3.6 E 6
DOB_5 (kg)	Water	6.4	1.6	5135
Calcium (kg)		15.2	6.24	5,247
Chloride (kg)		43.7	17.3	15,713
COD (kg)		8.2	2.3	5,254
Dissolved solids (kg)		4.4	4.74	301
Unspecified oils (kg)		4.8	6.6	1,810
Silicon (kg)		7.7	11	1,110
Sodium (kg)		9.8	8.8	8,201
Sulfate (kg)		14.9	8	8,800
Calcium	Soil	0.07	0.02	49.3
Chloride (kg)		0.4	0.05	46.3
Unspecified oils (kg)		2.1	0.43	1,916
Solid waste (kg)		15.7	1494	11,972
Removed diesel (m^3)		0.333	60.68	213.03

substance in these categories was unspecified oils released during diesel, electricity and asphalt production.

An analysis of **Fig. 3**, which presents the comparison of average normalized primary and secondary impacts for each technology throughout the years, reveals a decrease in ANSIs noticeable in year 4 for the biopile treatment. This is mainly due to the recycling of asphalt at the site closure stage. Indeed, an environmental credit is awarded because new asphalt production is avoided by recycling. Comparison of the average normalized primary impacts (ANPIs) shows no significant decrease for NA, but a linear decrease is observed for both bioventing and biopile treatment. The final ANPI for bioventing (7.5e5 pers.year) is lower than that obtained for biopile treatment (2.08 e6 pers.year). This can be explained by the fact that the final diesel content is higher in soils treated with biopiles (while both technologies reached the same final concentration, a more significant volume of soil had to be treated using biopiles). Compared on an average value, the impacts from the residual contamination were more significant than the secondary impacts even at the end of the treatment for each technology.

2.3 Comparison of groundwater technologies

The groundwater technologies compared were pump and treat (PT), biosparging, chemical oxidation and natural attenuation (Nag). The treatment time necessary to reach regulation criteria differs considerably between the four technologies. PT requires 3 centuries, biosparging 36 years, Nag 6 years and chemical oxidation only 4 years.

The comparison of these technologies' input materials (**Table 7**) shows that the more a technology requires wells, the more diesel it requires for the machinery. In addition, results show that the longer the technology is used, the more electricity it consumes. PT uses activated carbon (531 ton) to treat the water while the others don't need water treatment system because no groundwater is withdrawn. The

Table 6: Total average normalized impacts by impact categories for the soil treatment stage

Categories (person*year)	Media	Natural attenuation	Bioventing	Excavation & biopiles
Global warming	Air	0.003	0.003	0.19
Ozone depletion	Air	0.02	0.004	9.53
Acidification	Air	0.42	0.31	27.2
Eutrophication	Air	0.6	0.57	29.3
	Water	0.38	0.12	3.0 E 2
	Soil	0.002	4.3 E-4	1.37
Photochemical smog	Air	0.52	0.48	30.3
Ecotoxicity	Air	0.22	0.22	23.5
	Water	**9.8 E 2**	**1.3 E 3**	**3.6 E 5**
	Soil	**4.02 E 2**	**89.2**	**3.8 E 5**
Human health cancer	Air	2.4 E-8	2.1 E-8	9.4 E-6
	Water	3.2 E-6	4.4 E-6	1.2 E-3
	Soil	1.4 E-6	3.0 E-7	0.001
Human health criteria	Air	1.04	0.76	54.2
Human health non cancer	Air	0.09	0.07	32.4
	Water	1.2 E-3	8.9 E-4	0.27
	Soil	2.0 E-4	3.8 E-5	0.15

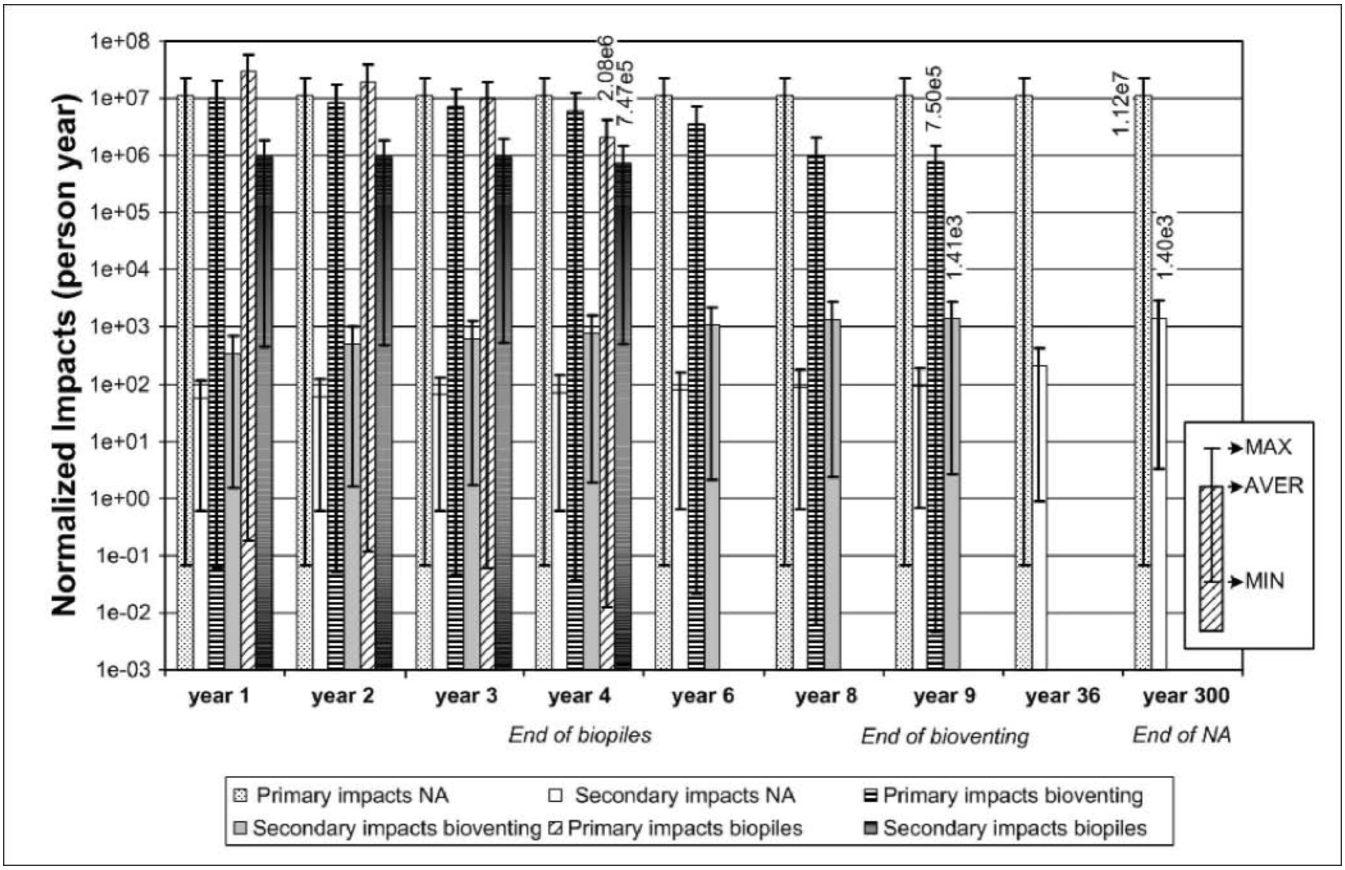

Fig. 3: Comparison of the normalized primary and secondary impacts for the soil treatment technologies over the years; the values indicate the average normalized impacts generated upon completion of treatment

analysis of the output emissions showed that when more transports were needed for a technology, its fossil CO_2 emissions were higher. Recycling diesel during the PT restrained the dissolved solids to end up in water emissions (–574.5 kg). Also, the chemical oxidation presents the most emissions in air, water and soil. This is mainly due to oxidant production and transport.

Comparing the four technologies according to their ANSI (**Table 8**) for each impact categories shows that chemical oxidation produces more impacts in all categories except for soil eutrophication for which PT has the most (2.67 pers.year vs 2.54 pers.year). This category is affected by phosphorous emissions, activated carbon production being its major source. Once again, the most important categories are water and soil ecotoxicity. The substance which had the most significant effect on these categories is unspecified oil released during diesel and electricity production.

Fig. 4 shows that the ANPIs are lower than the ANSIs in the last year of treatment for PT, biosparging and chemical oxidation. This is due to the fact that the remediation target was set at the detectable limit of C_{10}-C_{50} in water. The Nag has more ANPIs than ANSIs because when it reached the river, the residual concentration of hydrocarbon in the groundwater was 1 mgL^{-1} and the primary impacts associated with this concentration represents a high value. Nevertheless, this is an acceptable level according to the QRCS (criteria for diesel in surface water: 2.8 mgL^{-1} [30]). Note that even if chemical oxidation showed a very rapid decontamination efficiency, the associated environmental load was tremendous (1.26e6 pers.year) compared to the others (PT: 3.96e5 pers.year, Biosparging: 2.88e4 pers.year, Nag: 6.23e3 pers.year).

2.4 In situ technologies versus ex situ technologies

The in situ technologies studied to treat the vadose zone were natural attenuation and bioventing. The ex situ treatment studied was biopile. According to their primary and secondary impacts and their treatment time, natural attenuation was not a good option. After 300 years, the soil remains contaminated with a diesel concentration of 9,878 $mgkg^{-1}$, which represents a long-term contamination source for groundwater and eventually, surface water. Bioventing is a good option with only 8 years of treatment time. This is a reasonable time if the owner means to retain his proprietary rights for as long as needed. This treatment generated only few impacts and did not disturb the site. Biopile treatment achieved the same level of remediation as bioventing in only four years but with more than 500 times the secondary impacts (refer to Fig. 3). It also disturbed the site with the creation of a possibly hazardous hole and a large paved area. In this case, the in situ bioventing technology is the best for the environment. It allows obtaining the same results in a reasonable time and with little installations. The in situ groundwater treatment technology is characterized by the

Table 7: Mass input materials and mass output emissions for the groundwater treatment phase

Materials (inputs)		Pump & Treat	Biosparging	Chemical Oxidation	Natural Attenuation
Diesel $_{machinery}$ (m^3)		2.5	8.7	12.5	0.6
Diesel $_{transport}$ (m^3)		79.8	1.5	268.5	0.7
Cement (kg)		210.1	4,320.5	5,117	336
Bentonite (kg)		843.4	681	1,676	660
Sand (kg)		1,036	844	352	1,407
Gravel (kg)		–	–	2,076	–
PVC (kg)		459.1	1,814	513	257
Steel (kg)		–	–	10,534	–
Activated carbon (kg)		530,700	–	–	–
Electricity (GJ)		67,010	951	166	–
Clean soil (kg)		2,245	6,413	10,153	458
Latex (kg)		52.8	15	1.7	2
Glass (kg)		6,264	1,503	167	188
CH_3COOH (kg)		–	–	2,280	–
H_2O_2 (t)		–	–	4,406	–
Fe_2SO_4 (t)		–	–	9,844	–
Distilled water (t)		–	–	9,844	–
Emissions (outputs)					
Biogenic CO_2 (kg)	Air	4.1 E 5	1,690.3	3.5 E 5	19
Fossil CO_2 (kg)		6.8 E 5	44,769	6.56 E 6	4,817
Aluminum (kg)	Water	458	7.8	19,842	0.22
Calcium (kg)		1,889	37	18,689	1.7
Chloride (kg)		1,752.4	140	29,917	18
COD (kg)		306	14.4	17,970	2.1
Dissolved solids (kg)		–574.5	51	3,557	6.3
Unspecified oils (kg)		1,896	139	4,821	9.2
Silicon (kg)		4,082	64	42,269	1.2
Sodium (kg)		540	71.2	12,802	10
Sulfate (kg)		2,657	48.7	20,848	1.8
Calcium	Soil	36.5	0.14	70.5	0.02
Chloride (kg)		21.5	0.8	126	0.06
Unspecified oils (kg)		78.6	4.2	1,433	1.4
Solid waste (kg)		5.4 E 5	2,072	15,343	277.4
Removed diesel (m^3)		246.6	165.02	165	4.964

Table 8: Total average normalized impacts by impact categories for the groundwater treatment stage

Categories (person*year)	Media	Pump & Treat	Biosparging	Chemical Oxidation	Natural Attenuation
Global warming	Air	0.57	0.025	3.67	0.003
Ozone depletion	Air	1.07	0.026	27.1	0.003
Acidification	Air	30.1	2.76	2.5 E 2	0.34
Eutrophication	Air	45.7	4.84	2.7 E 2	0.65
	Water	20.2	0.69	6.6 E 2	0.08
	Soil	2.67	4.3 E-3	2.54	4.7 E-4
Photochemical smog	Air	85.7	4.88	2.4 E 2	0.61
Ecotoxicity	Air	31.6	1.44	4.9 E 2	0.13
	Water	**3.8 E 5**	**2.8 E 4**	**9.7 E 5**	**1.9 E 3**
	Soil	**1.6 E 4**	**8.5 E 2**	**2.9 E 5**	**1.1 E 2**
Human health cancer	Air	7.0 E-6	1.1 E-7	8.2 E-5	4.1 E-9
	Water	0.001	9.2 E-5	0.003	6.1 E-6
	Soil	5.2 E -5	2.8 E-6	9.5 E-4	3.5 E-7
Human health criteria	Air	2.2 E 2	8.78	6.1 E 2	0.7
Human health non cancer	Air	24.3	0.38	2.9 E 2	0.014
	Water	0.21	0.013	5.1	9.4 E-4
	Soil	7.1 E -3	4.0 E-4	0.13	4.8 E-5

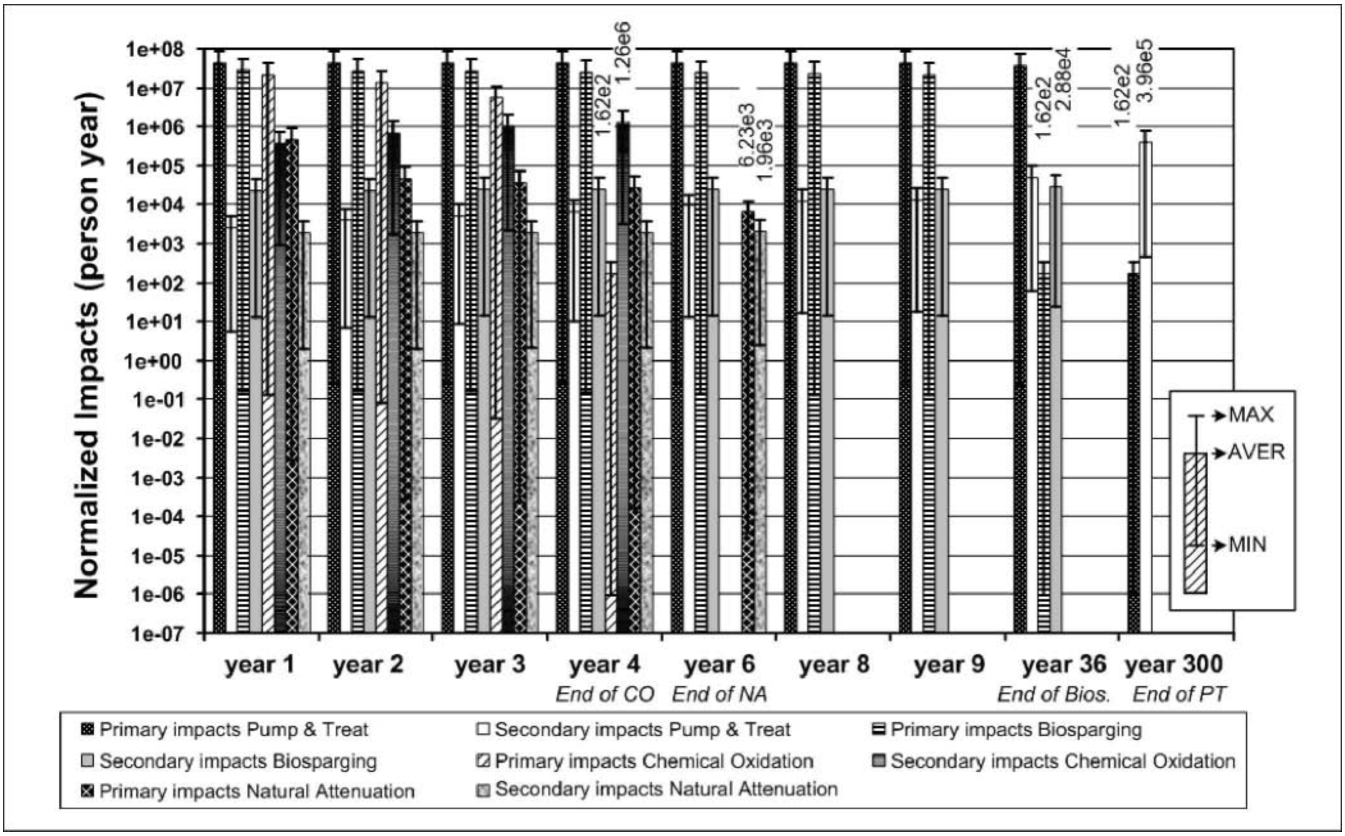

Fig. 4: Comparison of the normalized primary and secondary impacts for the groundwater treatment technologies over the years; the values indicate the average normalized impacts generated upon completion of treatment

water being treated inside the aquifer whereas the ex situ treatment of water is conducted outside. In this case, PT is the only technology to be considered as ex situ. All 4 technologies reached, with different treatment times, the regulated remediation criteria or better. The chemical oxidation provided the same results as PT and biosparging with 3 and 43 times more secondary impacts, respectively (refer to Fig. 4). Even though they generate less secondary impacts, treatment durations for PT and biosparging (300 and 36 years, respectively) may be considered too long for a site owner. Natural attenuation of the plume cannot be compared to the other technologies because the contamination source had been removed prior to its application. In such a case, it should not be considered as a treatment technology but as a monitoring program. It should be noted that it is not because an in situ technology is used that the environmental load it generates is lighter.

2.5 Selection of the best scenario

Having compared all technologies for each treatment zone, the four organized scenarios of Table 1 were compared next. **Fig. 5** presents the normalized primary and secondary impacts for the four scenarios upon completion of treatment.

The first scenario reflects the reality of a site whose owner wants to retain his proprietary rights for as long as needed with minimum intervention on his part. According to the results obtained, a soil treatment is required in order to render this first scenario efficient. Even if this scenario has the second least secondary impacts (4e5 pers.year), environmentally, it is the worst (since the soil remains contaminated in the end). From an environmental point of view, the second scenario is the best, generating the least primary and secondary impacts. Complete remediation for this scenario requires, however, 38 years which can be considered significant for a time-stressed owner. The third scenario which involves chemical oxidation could be interesting because of its short treatment time (11 years) and high treatment efficiency. Its secondary impacts are the highest of all scenarios (1.3e6 pers.year). Also, the LCA did not take into account the technological risks that the staff may encounter. Explosions may occur during oxidant injection since the oxidizing reaction is exothermic. Moreover, the risk associated with the transport of chemicals was not taken into account in the LCA. Hence, the third scenario is a good option in terms of treatment time and efficiency but there are other considerations to keep in mind that were not included in the analysis. It could be a good option if experienced staff is hired and if chemicals can be purchased near the site to reduce the risks associated with long distance transport. The fourth scenario has the lowest treatment time with 8 years. However, the residual contamination remains higher than for scenarios 2 and 3, although it was controlled and maintained below the regulation criterion. For an owner, a shorter treatment time is often preferred. Nevertheless, this scenario requires a lot more preparation compared to the others. It demands more organization and a large area of land must be available, which is not always the case. Contrary to what

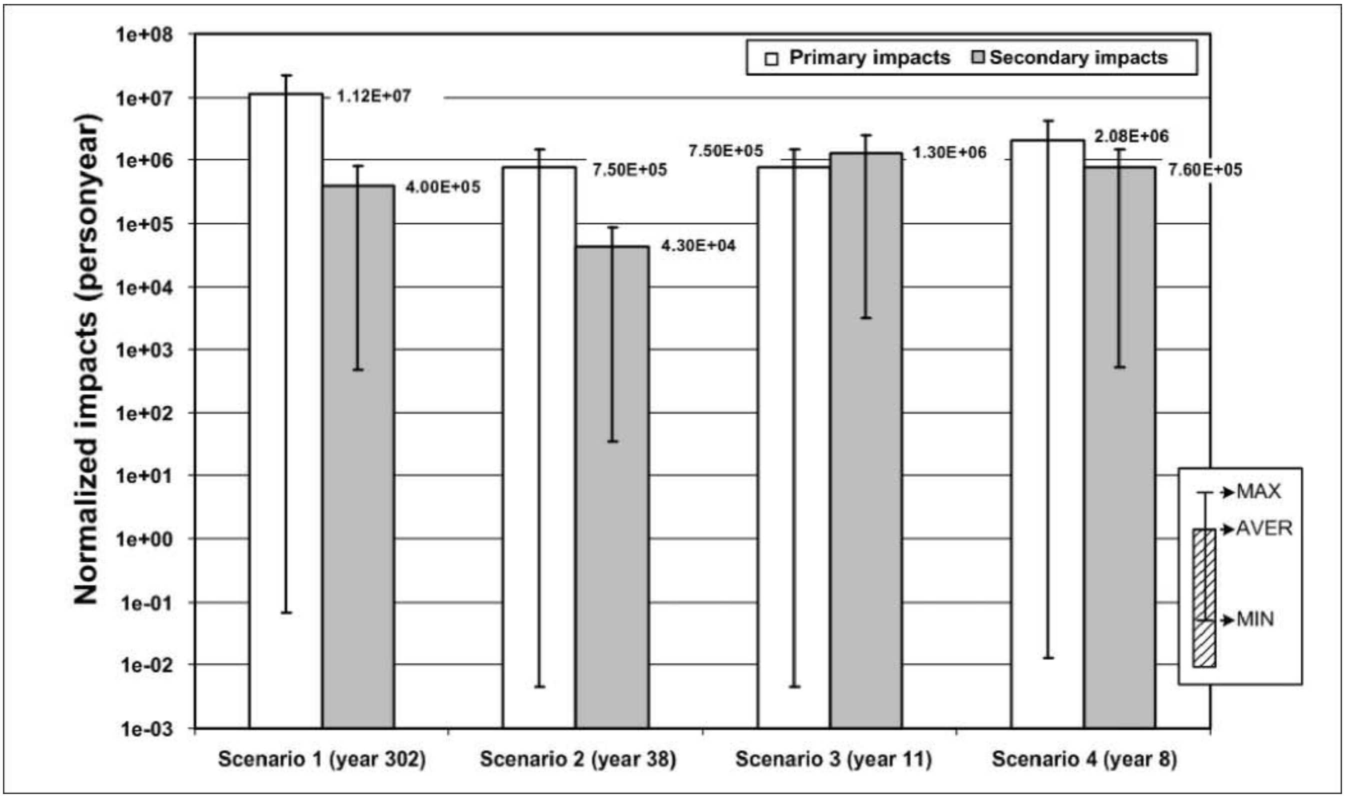

Fig. 5: Comparison of the normalized primary and secondary impacts for the four scenarios generated during their last year of treatment. The values indicate the average normalized impacts

could be expected, the secondary impacts of scenario 4 (7.6e5 pers.year) were lower than those for scenario 3. Since the best scenario selection will depend on the site owner's priorities, one could try to combine different technologies for the same zone. For example, pump and treat and biosparging could be used together to treat the groundwater faster or chemical oxidation (used with less wells) combined with natural attenuation to reduce the risk. Depending on his priorities, two scenarios can be proposed to a site owner. The first one considers low environmental impacts as a priority. The proposed scenario has low technological risks, but has a considerably long treatment time: it is scenario 2. This scenario is also probably the cheapest although no analysis has been conducted to confirm this. The second proposed scenario is based on the shortest treatment time. It combines the fastest technologies of scenarios 3 and 4. The proposed scenario would use bioslurping to remove the LNAPL phase, excavation and treatment of soils in biopiles and chemical oxidation for the residual contamination in groundwater. Even if less oxidant would be needed to remediate the groundwater the technological risk associated with this technology would still be present. Another drawback of this option is that it will add secondary impacts due to the transport of contaminated and clean soil. This option would also probably be the most expensive.

2.6 Limit of LCA

In this analysis, only average values of primary and secondary impacts were compared. The minimum and maximum results are so different that if the comparison is conducted solely on the minimum values, primary impacts are always smaller than secondary impacts (see Figs. 2 through 5). In Fig. 2, the differences between minimum values of NSI and NPI are only of one to two orders of magnitude. In Fig. 4, however, the difference can reach up to more than 8 orders of magnitude, the NSI taking precedence over NPI. The large variety of possible results for the primary impacts is due to only 3 impact categories that need min. and max. factors to characterize residual diesel in soil and water (whose factors are the same). The min. and max. factors used by these categories are: ecotox.: 0.0001653000; HHC: 0.554367–19048.49 and HHNC: 0.020207–842.2572. The secondary impacts have min. and max. values due to unspecified oils that have the same CF as diesel and to 6 other aggregated substances characterized in the photochemical smog category. It was important to include the unspecified oils in the secondary impacts because it was one of the most significant emissions (more than 5% of all emissions) for all scenarios. Importance should be given to primary impacts in a site remediation LCA but, more detailed CFs are needed to reflect a better reality of the residual contamination on a specific site. Since no CFs exist for aggregated substances in the US EPA TRACI method, this aspect requires improvement. It was important to use the North American characterization method to get a more representative geographical context since no Canadian equivalent was available.

3 Conclusions and Recommendations

A comparison of the environmental performance of four treatment scenarios has been conducted. Globally, the fast-

est scenario treated the site in 8 years while the slowest required three centuries and left the contamination source in place. The all biological in situ scenario (2) took 38 years to remediate the site compared to the biological/ chemical (3) which only required 11 years. Secondary and primary impacts of all scenarios were compared. The all biological in situ scenario (2) showed the least secondary and primary impacts of all four. On the other hand, the in situ biological/ chemical scenario (3) produced the most secondary impacts and quickly reduced primary impacts. Surprisingly, the ex situ scenario (4) generated twice the secondary impacts compared to the traditional one (1), which produced almost 10 times the secondary impacts of the all biological scenario (2). The most affected impact category was water ecotoxicity. Based on these results, two scenarios were proposed, one with low environmental impacts but with a long treatment time and another with short treatment time but with high environmental impacts. Finally, the LCA comparison of treatment scenarios for both soil and groundwater clearly gave a better idea of the best combination of technologies. Results showed that an in situ treatment scenario does not necessarily provide for a better environmental outcome than an ex situ treatment scenario. Also, additional work is needed to improve characterization factors of aggregated substances in order to improve the LCA's performance.

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